IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: Akihiko Itami	Examiner: Christopher D. Rodee
Serial No.　10/663,137	Art Unit:　　1756
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Mail Stop Amendment
Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

DECLARATION UNDER 37 CFR § 1.132

I, Mr. Akio Itami, declare as follows:

1. I graduated from Tsukuba University in March 1987, with a Master's Degree in Organic Chemistry.

2. Since April 1987, I have been employed by Konica Corporation (now Konica Minolta Business Technologies Inc.) and have been engaged in research and development of electrophotographic materials.

3. The experiment discussed in this Declaration was conducted under my supervision and control.

4. Experiment: Photoreceptors were prepared. "Sample G" is a copy of JP 1-065561 ("JP") Sample G, except that the CGL and CTL were applied on an aluminum substrate having an inter layer as described in the present application at pages 81-82. Development condition was the same as Example of the present application as described

1

at page 98, except that the initial object charging potential was set at -610 V. Toner lBk described at pages 90-91 of the present application was employed.

Sample G photosensitive layer contains nonmetal phthalocyanine compound CGM and developing condition with electrical field intensity of 32.1 V/ μm.

Sample G (Mod 1) is a modification of Sample G in which thickness of CGL is reduced to give electrical field intensity of 46.9 V/ μm, closed to claimed value but outside. This is a comparative sample.

Sample G (Mod 2) is a modification of Sample G in which thickness of CGL is reduced to give electrical field intensity of 50.8 V/ μm, close to the claimed value but outside. This is a comparative sample, since the CGM is phthalocyanine compound, but not N-type. This sample shows that even though the electric field satisfies the value as claimed in this application it does not give a sufficient black spot reducing effect in combination with phthalocyanine.

Sample G (Mod 3) - (Mod 4) each are a modification of Sample G in which thickness of CGL is reduced to give electrical field intensity of 50.8 V/ μm, and CGM is replaced by Perylene A, N-type one. These are inventive samples. Though the electrical field intensity is identical with (Mod 2), this sample gives satisfactory result in combination with N-type CGM, Perylene A.

Sample G (Mod 6) - (Mod 8) each are a modification of Sample G in which thickness of CGL is reduced to give electrical field intensity of 50.8 V/ μm, and CGM is replaced by Perylene A, N-type one. These are inventive samples.

The evaluation condition is the same as Example of the present application (page 97-102), measured in 50,000 th copies. This is different from JP evaluation condition, in which evaluation was conducted at the initial stage copy.

The result shows that the combination of specific electric field with an N-type CGM gives excellent effect used further in combination with specific toner.

2

	CGM	Thickness of CGL (µm)	Thickness of CTL (µm)	Thickness of photosensitive layer (µm)	Electrical field intensity E (V/µm)	Black spots	White spotting	Image density
Sample G* (Comparative)	Phthalocyanine**	9	10	19	32.1	C	C	B
Sample G (Mod 1) (Comparative)	Phthalocyanine	3.0	10	13.0	46.9	C	B	A
Sample G (Mod 2) (Comparative)	Phthalocyanine	2.0	10	12.0	50.8	C	B	A
Sample G (Mod 3) (Inventive)	Perylene A***	2.0	10	12.0	50.8	B	A	A
Sample G (Mod4) (Inventive)	Perylene A	1.0	11	12.0	50.8	A	A	A
Sample G (Mod 5) (Inventive)	Perylene A	0.5	11.5	12.0	50.8	A	A	A
Sample G (Mod 6) (Inventive)	Kawahara's CGM (4)****	2.0	10	12.0	50.8	B	A	A
Sample G (Mod 7) (Inventive)	Kawahara's CGM (4)	1.0	11	12.0	50.8	B	A	A
Sample G (Mod 8) (Inventive)	Kawahara's CGM (4)	0.5	11.5	12.0	50.8	A	A	A

*   Sample G: Described in JA 64-065561.

**   Phthalocyanine: Nonmetal phthalocyanine compound A used in Sample G (Preparation is disclosed in JA 64-065561).

***   Perylene A: Described in the present application at page 87.

****   Kawahara's CGM (4): Disclosed in Kawahara's column 13.

3

4. I further declare that all statements made herein of my own knowledge are true and that all statements made upon information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Executed on: _____   By: _____

Mr. Akio Itami

Partial Translation of JPA64-65561

Page 5, right lower column, line 6 to page 6, right upper column, line 10.

The photoreceptor used in the present invention is characterized in that a phthalocyanine compound is contained in the carrier generation layer. This improves the potential stability at the time of repeated use of the photoreceptor, and reduces the phenomenon of memory and residual potential, thereby ensuring stability. Moreover, since the phthalocyanine compound is highly sensitive in the long wavelength region, this arrangement provides a high-performance photoreceptor matched with the image forming method according to the reversal development wherein a semiconductor laser is used as a light source.

To be more specific, because the aforementioned phthalocyanine compound is highly sensitive to the semiconductor laser, and the thickness of the carrier generation layer is increased, a high degree of sensitiveness to light can be maintained even if the ratio of the content of the carrier generation material relative to that of the binder material is 1/2 or less.

For the same reason, the absorption coefficient κ of the carrier generation layer can be increased in the aforementioned Eq. (1). Thus, combined with the fact that the thickness d_{CGM} of the carrier generation layer is made as large as 1 μm or more, the value of Φ can be reduced, and therefore, the overall transmittance of the semiconductor laser beam can be further reduced, with the result that uneven density in the form of an interference fringe is less likely to occur.

In addition to the aforementioned characteristics, the present invention is further characterized in that the absolute value of the charging potential represented by $|V^H|$ is restricted to a particular range of $|V^H| = 500$ through 900V. To be more specific, when $|V^H| < 500$V, the required field intensity cannot be easily obtained. If $|V^H| > 900$V, the thickness of the photosensitive layer have to be increased, with the undesired result that the sensitivity is reduced.

The present invention is still further characterized in that the $|V^H| - |V^{DC}|$ as a difference between $|V^H|$ and $|V^{DC}|$ (absolute value of direct current bias) is specified at 0 through 200V. To be more specific, when $|V^H| - |V^{DC}| < 0$, fogging will occur. If $|V^H| - |V^{DC}| > 200$V, deposition of a carrier (in the case of a two-component developer) and deposition of toner of inverse polarity (in the case of a bipolar one-component

developer) will occur.

Accordingly, based on the present invention, $|V^H| = 500$ through 900V (preferably 550 through 7000V) and $|V^H| - |V^{DC}| = 0$ through 200V should be adopted. To carry out reversal development under these conditions is indispensable to ensure a high-quality image, free from black dots, characterized by a high degree of sensitivity.

Page 6, right lower column, line 15 to page 8, left upper column, line 14.

The photoreceptor used in the present invention, for example, the electrophotographic photoreceptor can be configured in various structures.

Figs. 1 and 2 illustrate a general structure.

In the photoreceptor shown in Fig. 1, the carrier generation layer 2 according to the present invention is arranged on the semiconductive substrate 1, and a carrier transport layer 4 is arranged thereon. A photosensitive layer 5 is composed of the carrier generation layer 2 and carrier transport layer 4. A carrier generation material 10 and a carrier transport material (mutually soluble with the binder material) are contained in the carrier generation layer 2.

In the photoreceptor of Fig. 2, an intermediate layer or undercoating layer 3 is arranged between the semiconductive substrate 1 and photosensitive layer 5, and is assigned with a function as the adhesive layer and others. The film thickness of the layer 3 is in the range from 0.03 through 20 μm.

In the photoreceptor shown in Figs. 1 and 2, an intermediate layer having a blocking function can be arranged between the carrier generation layer and carrier transport layer. Further, to prolong the press life, a protective layer (protective film) may be provided on the photoreceptor surface. For example, synthetic resin coating maybe provided.

The ratio of the content of the carrier generation material in the carrier generation layer relative to that of the binder material should be 1/2 or more, but is preferably 1/3 through 1/20, and more preferably 1/4 through 1/10. The ratio of the carrier generation material content is greater than that, black dots and others will appear conspicuously or will appear easily. However, if the percentage of the carrier generation material is too small, the sensitiveness to light and others will be reduced.

The film thickness of the carrier generation layer is 1 μm or more. It is preferably 2 μm or more, and more preferably 5 through 25. If the film thickness is smaller than the above level, carrier injection cannot be prevented, or cannot be

prevented easily. However, if the film thickness is too great, the light carrier have to travel a long distance, with the result that a sufficient transport capacity cannot be obtained in general cases. Thus, residual potential tends to rise easily in the case of repeated use. The film thickness of the carrier generation layer is preferably 3/4 or less that f the overall photosensitive layer. If this film thickness ratio is greater than the aforementioned level, the charging potential tends to decline.

The film thickness ratio between the carrier generation layer and carrier transport layer is preferably in the range of (film thickness of the carrier generation layer: film thickness of the carrier transport layer) = (1: 20) through (1: 1).

The film thickness of the carrier generation layer is preferably 2 μm or more. If the film thickness is less than 2 μm, the surface of the carrier transport layer is subjected to mechanical damage at the time of repeated use depending on the mode of operation such as development and cleaning. Part of the layer may be removed, and a black stripe may appear on the image.

The film thickness of the photosensitive layer as a whole is preferably in the range from 10 through 40 μm, and more preferably in the range from 15 through 30 μm. If this film thickness is smaller than the aforementioned level, a charging potential will be reduced due to the excessively thin layer, and the press life will also be reduced. Further, if the film thickness is greater than the aforementioned level, the residual potential will rise on the contrary. Not only that, there will be the similar phenomenon as that in the aforementioned case where the carrier generation layer is too thick. Thus, a sufficient transport capacity cannot be ensured, and residual potential tends to rise at the time of repeated use.

The content of the carrier transport material in the carrier generation layer is preferably 1 through 100 relative to 100 parts by weight of the binder material, and is more preferably 5 through 50 parts by weight. If the content of the carrier transport material is greater than this level, film strength tends to be reduced. If the content of the carrier transport material is smaller than this level, the carrier mobility in the CGL will be reduced and residual potential will rise. Further, sensitiveness to light will decline, with the result that a faulty image tends to be produced.

The ratio of the content of the carrier generation material relative to that of the carrier transport material is preferably carrier generation material : carrier transport material) = (1 : 100) through (5 : 1), and more preferably (1 : 10) through (1 : 1) in terms of weight ratio in order to ensure an effective performance of functions of each of them.

When a granular carrier generation material is dispersed to form a

3

photoconductive layer, this carrier generation material is preferably composed of granules having an average particle size of 0.1 μm or more without exceeding 5 μm or preferably 0.2 μm or more without exceeding 2 μm. To be more specific, if the particle size is too large, a poor dispersion into the layer will result. If the particle size is too small, condensation tends to occur. This will result in increased layer resistance, severer crystal defects, lower sensitivity, reduced repeatability and poorer charging capacity. Further, this will impose a limit to the further efforts for microstructure.

The aforementioned phthalocyanine (Pe) is characterized as follows:

Phthalocyanine as one of the organic photoconductive materials is characterized in that the photoconductive range is expanded into the long wavelength region, as compared to other materials. This characteristic is widely known. In the process wherein the α type phthalocyanine is converted into stable crystal type β phthalocyanine, various crystal types of phthalocyanine have been observed. These photoconductive phthalocyanine compounds include the X-type non-metallic phthalocyanine disclosed in the Official Gazette of Japanese Patent Tokkosho 49-4338, and τ, τ', η, and η'-type non-metallic phthalocyanine compounds disclosed in the Official Gazette of Japanese Patent Tokkaisho 58-182639 and Official Gazette of Japanese Patent Tokkaisho 60-19151.

Page 16, left lower column, line 9 to right lower column line 12.

<Synthesis of phthalocyanine compound>

Prior to introducing the embodiment, the following describes an example of synthesizing the non-metallic phthalocyanine compound A having the characteristics shown in Figs. 7 and an example of synthesizing the τ type non-metallic phthalocyanine compound.

(Example of synthesis)

50 g of lithium phthalocyanine was added into 600 ml of concentrated sulfuric acid sufficiently agitated at 0 degrees Celsius. Then the resultant mixture was agitated at the same temperature for two hours. The resultant solution was then filtered through a glass-made coarsely sintered funnel. It was gradually poured into 4 liters of ice and water while being agitated. After having been left to stand for several hours, the mixture was filtered and the lump having been obtained was washed in water until it becomes neutral. Then the lump was washed several times in methanol in the final stage and was dried in open air. The powder obtained by drying was

4

extracted by acetone on a continuous basis in an extractor, and was turned into blue powder.

In the above step, lithium was repeatedly subjected to precipitation in order to assure the residue of salt. In the aforementioned manner, 30.5 g of blue powder was obtained. This product has been found out to be matched with the graphic representation of the α type phthalocyanine compound by X-ray diffraction.

Page 17, right upper column, line 1 to page 6, right upper column line 10.

<Preparation of photoreceptor>

The photoreceptors A through L in the embodiment and photoreceptors a through k in comparative examples were manufactured in the manner shown below wherein these photoreceptors were manufactured in the same production procedure.

20 g of the carrier generation material following were crushed at 40 rpm by a magnetic ball for 18 hours. After that, the solution obtained by dissolving a predetermined amount of polycarbonate resin [Panlite L1250] (by Teijin Kasei Co., Ltd.) in 1,000 mL of 1, 2-dichloroethane was added thereto, and the resultant mixture was dispersed for another 24 hours, thereby preparing a coating solution for the CGL (carrier transport layer) having a predetermined P/B ratio (the ratio of the content of the carrier generation material relative to that of the binder material (carrier generation material /binder material): this applies to the description hereinafter)).

It should be noted that, for the G and H in the embodiment, 10 g of the CTM (carrier transport material) was added to the coating solution for the CGL.

Then using a doctor blade, the aforementioned coating solution for CGL was applied onto the conductive support member composed of the polyethylene terephthalate having a thickness of about 75 μm produced by vapor deposition of aluminum. Thus, the carrier generation layer having a predetermined thickness of film was formed.

Further, 11.25 g of a predetermined carrier transport material and 15 g of a predetermined binder were dissolved in 100 mL of 1, 2-dichloroethane, and the resultant solvent was coated on the carrier generation layer using a doctor blade. This was dried at 90 degrees Celsius for one hour to form a carrier transport layer.

Here in the photoreceptors G and H, the carrier transport substance was added to the carrier generation layer.

In each of the photoreceptors, the carrier transport substance in the coating

solution was diffused into the carrier generation layer at the time of formation of the carrier transport layer. Thus, the carrier transport substance was contained in the carrier generation layer. (However, the photoreceptor d was not subjected to such diffusion. No carrier transport substance was contained in the carrier generation layer.)

As described above, the photoreceptors A through L and a through k, each having a separate structure and prescription, were manufactured according to the common production procedure.

To be more specific, the amount of resin contained in the carrier generation layer, P/B ratio and film thickness; and the film thickness of the carrier generation material, the carrier transport material used in the carrier transport layer, binder material and photoreceptor are varied for each of the photoreceptors.

The structure and prescription of each photoreceptor are given in the following Table 2:

Table 2 (in part)

		CGL				CTL		Photosensitive layer
	(1)	(2)	P/B ratio	(3)	CGM	CTM	Binder	Thickness
Example	I	100	1/5	6	[I]	[III]	[Y]	21
	J	100	1/5	7	[I]	[III]	[Y]	22
	K	100	1/5	11	[I]	[III]	[Y]	24
	L	100	1/5	16	[I]	[III]	[Y]	27
	G	160	1/8	9	[I']	[VI]	[Y]	19
	H	160	1/8	17	[I']	[VI]	[Y]	27
	D	60	1/3	2	[I']	[VI]	[Z]	26
	E	60	1/3	6	[I']	[VI]	[Z]	23
	F	60	1/3	13	[I']	[VI]	[Z]	20
	A	40	1/2	1	[I']	[III]	[Z]	25
	B	40	1/2	7	[I']	[III]	[Z]	22
	C	40	1/2	1.2	[I']	[III]	[Z]	21

(1): Photoreceptor   (2): Resin content   (3): Thickness

* No carrier generation material contained in the carrier generation layer.

The binder in the carrier generation layer is composed of polyvinyl butyral (which does not dissolve in the carrier transport layer).
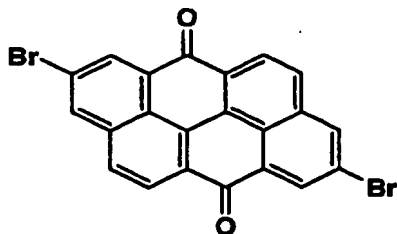
Tetrahydrofuran is used as a solvent

Carrier generation material

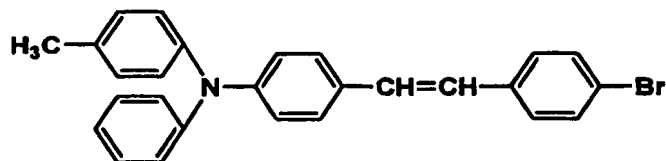[I]: Aforementioned non-metallic phthalocyanine compound A

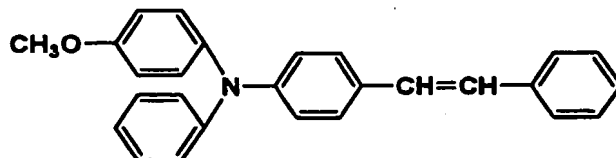[I']: Aforementioned □ type phthalocyanine compound A

[II]:

H₃C appears...

[Chemical structure II: a polycyclic dibromo-dione]

Carrier transport material

[III]

[Chemical structure III]

[IV]

[Chemical structure IV]

Binder material [Y]

Polycarbonate resin [Panlite K-1300]

(by Teijin Kasei Co., Ltd.)

Binder material [Z]

Acryl resin [Dianal BR-85]

(by Mitsubishi Rayon Co., Ltd.)

Example 1 and Comparative example 1

Each of the twenty-one photoreceptors composed of photoreceptors A through L in the embodiment and photoreceptors a through k in comparative examples was placed on a modified version of the KONICA LP 3005 (by Konishiroku Photo Industry

Co., Ltd.)(LD-mounted printer). The grid voltage was adjusted so that the VH would be -600 +- 10 [V], and reversal development was carried out at a development bias of 480V. The black spot on the white ground of the copied image, the image density Dmax of the black portion (corresponding to the white portion of the original image) and moire fringe pattern were evaluated.

To evaluate the black spot, an image analyzer "Omnicon" (by Shimadzu Seisakusho Ltd.) was used to measure the particle diameter and the number of the black spots. Evaluation was made by checking the number of black spots per square centimeter each having a diameter of 0.05 mm or more.

Table 3 shows the criteria for black spots:

Table 3

Black spot of 0.05 mm or more in diameter	Evaluation
0/cm^2	A
1 through 3/cm^2	B
4 through 10/cm^2	C
11/cm^2 or more	D

A product can be put to practical use if the result of black spot evaluation is A, B or C, and cannot be used in the case of D. "B" is assigned to the Dmax of 1.3 or more, and "D" to the Dmax below 1.3. For moire fringe pattern, "B" indicates that the product is free of moire fringe pattern, and "D" indicates that a moire fringe pattern is found on the product.

Table 4 shows the Dmax and the presence or absence of moire fringe pattern based on the evaluation of the black spot for each photoreceptor.

8